IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of Koji MASAKI
Application No.: 10/598,842
Filed: September 13, 2006
For: RUBBER COMPOSITION AND PNEUMATIC TIRE USING THE SAME
Group Art Unit: 1764
Examiner: Irina Krylova
Confirmation No.: 1122

DECLARATION UNDER 37 C.F.R. § 1.132

I, Eiju Suzuki, declare that:

I am a co-worker of Mr. Koji Masaki who is the inventor of the above-captioned patent application.

I received my Master of Science and Technology from Keio University in 2002, and have been employed by Bridgestone Corporation since 2002, where I have been engaged mainly in research and development of new polymers.

I have made the following experiments in order to evaluate processability, storage modulus (G'), loss factor (tan δ) and fracture strength (TB) of:

(1) a rubber composition comprising an aromatic vinyl compound-diene compound copolymer (B) having a weight average molecular weight of more than 50,000 but not more than 300,000 and a styrene-butadiene copolymer (C) having a weight average molecular weight of not less than 300,000, as a working example of the present invention;

(2) a rubber composition comprising an aromatic vinyl compound-diene compound copolymer (B) having a weight average molecular weight of less than 50,000 and a styrene-butadiene copolymer (C) having a weight average molecular weight of not less than 300,000, as a comparative example;

(3) a rubber composition comprising an aromatic vinyl compound-diene compound copolymer (B) having a weight average molecular weight of more than 50,000 but not more than 300,000 and a styrene-butadiene copolymer (C) having a weight average molecular weight of less than 300,000, as a comparative example; and

(4) a rubber composition comprising an aromatic vinyl compound-diene compound copolymer (B) having a weight average molecular weight of less than 50,000 and a styrene-butadiene copolymer (C) having a weight average molecular weight of less than 300,000, as a comparative example.

<u>Experimental Procedure</u>

<Production method of SBR**>

Into a pressure glass vessel of 800 mL dried and purged with nitrogen are charged a cyclohexane solution of butadiene (16%) and a cyclohexane solution of styrene (21%) so as to so as to be 40 g of butadiene monomer and 10 g of styrene monomer, and 0.18 mmol of 2,2-ditetrahydrofuryl propane is charged thereinto, and 0.35 mmol of n-butyllithium (BuLi) is added thereto, and thereafter the polymerization is carried out in a warm bath at 50°C for 1.5 hours. The polymerization conversion is approximately 100%.

Then, 0.5 mL of a solution of 5 wt% of 2,6-di-t-butyl-p-cresol (BHT) in isopropanol is further added to the polymerization system to stop the reaction. Thereafter, the reaction mass is dried according to the usual manner to obtain a polymer (SBR**).

(Comparative Example A)

A rubber composition is prepared according to a compounding recipe shown in Table A by using:

the styrene-butadiene rubber having a weight average molecular weight of 25,000, comprising 25 mass% of styrene and having a vinyl bond content in butadiene portion of 65 mass%, which is the same as in the original Example 1, and;

the styrene-butadiene rubber (SBR**) having a weight average molecular weight of 250,000, comprising 25 mass% of styrene and having a vinyl bond content in butadiene portion of 60 mass%.

(Comparative Example B)

A rubber composition is prepared according to a compounding recipe shown in Table A by using:

the styrene-butadiene rubber having a weight average molecular weight of 25,000, comprising 25 mass% of styrene and having a vinyl bond content in butadiene portion of 65 mass%, which is the same as in the original Example 1, and;

the styrene-butadiene rubber (#1500, made by JSR Corporation) having a weight average molecular weight of 450,000, comprising 23.5 mass% of styrene and having a vinyl bond content in butadiene portion of 18 mass%.

(Comparative Example C)

A rubber composition is prepared according to a compounding recipe shown in Table A by using:

the styrene-butadiene rubber having a weight average molecular weight of 80,000, comprising 25 mass% of styrene and having a vinyl bond content in butadiene portion of 65 mass%, which is the same as in Example 3, and;

the styrene-butadiene rubber (SBR**) having a weight average molecular weight of 250,000, comprising 25 mass% of styrene and having a vinyl bond content in butadiene portion of 60 mass%.

(Comparative Example D)

A rubber composition is prepared according to a compounding recipe shown in Table A by using:

the styrene-butadiene rubber having a weight average molecular weight of 120,000, comprising 25 mass% of styrene and having a vinyl bond content in butadiene portion of 65 mass%, which is the same as in Example 4, and;

the styrene-butadiene rubber (SBR**) having a weight average molecular weight of 250,000, comprising 25 mass% of styrene and having a vinyl bond content in butadiene portion of 60 mass%.

<Evaluation>

With respect to the rubber compositions of the above-described Comparative Examples A-D, as well as Comparative Example 1 and Examples 3 and 4 shown in the originally filed specification, the processability, storage modulus (G'), loss factor (tan δ) and fracture strength (TB) are evaluated. The results are shown in Table B.

The processability is evaluated by an index on the basis that Comparative Example 1 disclosed in the originally filed specification is 100 by measuring a Mooney viscosity ($ML_{1+4}/130°C$) of the rubber composition at 130°C according to JIS K6300-1994. The smaller the index value, the better the processability.

The storage modulus and loss factor are evaluated by an index on the basis

that Comparative Example 1 disclosed in the originally filed specification is 100 by measuring G' value and tan δ by means of a low heat-buildup viscoelasticity measuring apparatus (made by Rheometrix Corp.) under conditions that a temperature is 50°C and a strain is 5% and a frequency is 15 Hz.

The fracture strength (TB) is measured according to JIS K6301-1995. It is evaluated by an index on the basis that Comparative Example 1 disclosed in the originally filed specification is 100. The larger the index value, the better the fracture strength.

Table A

Compounding	parts by mass
Copolymer (C) *6	100
C/B *2	65
Stearic acid	2
Zinc oxide	3
Antioxidant *3	1
Vulcanization accelerator *4	0.4
Vulcanization accelerator *5	1
Sulfur	1.75
Copolymer (B)	30

*6: SBR #1500 (made by JSR Corporation) or;
the above-described SBR**

*2: ISAF, Seast 3H, made by Tokai Carbon Co., Ltd.

*3: Nocrac 6C

*4: Nocceler D

*5: Nocceler NS

Table B

		Comparative Example 1	Comparative Example A	Comparative Example B	Comparative Example C	Comparative Example D	Example 3	Example 4
Copolymer (B)	kind St/Vi	(aromatic oil) -	SBR 25/65	SBR 25/65	SBR 25/65	SBR 25/65	SBR 25/65	SBR 25/65
	Molecular weight (Mw)	-	25,000	25,000	80,000	120,000	80,000	120,000
Copolymer (C)	kind St/Vi	#1500 23.5/18	SBR** 25/60	#1500 23.5/18	SBR** 25/60	SBR** 25/60	#1500 23.5/18	#1500 23.5/18
	Molecular weight (Mw)	450,000	250,000	450,000	250,000	250,000	450,000	450,000
Processability	ML_{1+4} (index)	100	81	87	92	95	97	102
Storage modulus	G' (index)	100	105	107	112	117	118	125
Loss factor	tan δ (index)	100	97	95	87	83	82	77
Fracture strength	TB (index)	100	93	102	96	97	126	133

<Summary>

As seen from the results of Comparative Example A and Examples 3 and 4, (4) the rubber composition comprising the copolymer (B) having a Mw of less than 50,000 and the copolymer (C) having a Mw of less than 300,000 has a much lower storage modulus (G'), a much higher loss factor (tan δ) and a much lower fracture strength (TB) as compared to (1) the rubber compositions comprising the copolymer (B) having a Mw of more than 50,000 but not more than 300,000 and the copolymer (C) having a Mw of not less than 300,000.

Further, as seen from the results of Comparative Example B and Examples 3 and 4, (2) the rubber composition comprising the copolymer (B) having a Mw of less than 50,000 and the copolymer (C) having a Mw of not less than 300,000 has a much lower storage modulus (G'), a much higher loss factor (tan δ) and a much lower fracture strength (TB) as compared to (1) the rubber compositions comprising the copolymer (B) having a Mw of more than 50,000 but not more than 300,000 and the copolymer (C) having a Mw of not less than 300,000.

Furthermore, as seen from the results of Comparative Examples C and D and Examples 3 and 4, (3) the rubber compositions comprising the copolymer (B) having a Mw of more than 50,000 but not more than 300,000 and the copolymer (C) having a Mw of less than 300,000 have a lower storage modulus (G'), a higher loss factor (tan δ) and a much lower fracture strength (TB) as compared to (1) the rubber compositions comprising the copolymer (B) having a Mw of more than 50,000 but not more than 300,000 and the copolymer (C) having a Mw of not less than 300,000.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under § 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 4 - 8 - 2011. Declarant: ______

Eiju Suzuki